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### ***published in***

Monthly Notices of the Royal Astronomical Society: Letters  
2008

### ***DOI (link to publisher)***

[10.1111/j.1745-3933.2008.00507.x](https://doi.org/10.1111/j.1745-3933.2008.00507.x)

### ***document version***

Publisher's PDF, also known as Version of record

### [Link to publication in VU Research Portal](#)

### ***citation for published version (APA)***

Ivanov, T. I., Salumbides, E. J., Vieitez, M. O., Cacciani, P. C., de Lange, C. A., & Ubachs, W. M. G. (2008). Extreme-ultraviolet laser metrology of OI transitions. *Monthly Notices of the Royal Astronomical Society: Letters*, 389(1), L4-L7. <https://doi.org/10.1111/j.1745-3933.2008.00507.x>

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# Extreme-ultraviolet laser metrology of O I transitions

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Accepted 2008 May 28. Received 2008 May 23

## ABSTRACT

Some 16 transitions in atomic oxygen originating from its <sup>3</sup>P ground state were measured using a tunable narrow-band extreme-ultraviolet laser source, with an unprecedented accuracy of  $\Delta\lambda/\lambda = 8 \times 10^{-8}$ . The results are relevant for comparisons with spectral absorption features observed in the line of sight of quasars, in order to test a possible variation of the fine-structure constant  $\alpha$  on a cosmological time-scale.

**Key words:** atomic data – methods: laboratory – techniques: spectroscopic – quasars: absorption lines – ultraviolet: general.

## 1 INTRODUCTION

The search for a variation in fundamental constants on a cosmological time-scale of  $\sim 10^{10}$  yr is a topic of strong interest. The method employed is based on a comparison between spectral absorption lines at high redshift, observed in the line of sight of quasar systems, with the same transitions observed at zero redshift in the laboratory. Currently, two different approaches are pursued. One is the search for a variation in the dimensionless proton–electron mass ratio  $\mu$ , which employs transitions associated with the Lyman and Werner band of molecular hydrogen (Reinhold et al. 2006), its isotopologue HD (Ivanov et al. 2008) and the inversion splitting in ammonia (Flambaum & Kozlov 2007). The other addresses the fine-structure constant  $\alpha = e^2/(4\pi\epsilon_0\hbar c)$ , and is based on a comparison between atomic and ionic spectral absorption lines (Webb et al. 1999, 2001) that all depend in a different way on a possible variation in  $\alpha$ . Various spectral transitions have been included in a comprehensive analysis, dubbed the many multiplet (MM) method (Dzuba, Flambaum & Webb 1999). The astrophysically observed transitions falling in the atmospheric transmission window of visible and near-ultraviolet wavelengths correspond to laboratory wavelengths (i.e. zero redshift) in the deep-ultraviolet and extreme-ultraviolet (XUV) wavelength ranges, if redshifts of  $z > 2$  are probed. The implementation of the MM method is most effective if transitions associated with various elements at different degrees of ionization, covering a broad dynamic range of absorption strengths, are probed in the same highly redshifted absorbing clouds in the line of sight of quasars. For the purpose of such a comparison, a list of transitions that require improved laboratory wavelength metrology has been identified (Berengut et al. 2006).

In a program to provide accurate laboratory measurements of relevant spectral transitions in the XUV wavelength range for the pur-

pose of comparison with quasar absorption data, we have previously investigated transitions in C I (Labazan et al. 2005), N I (Salumbides et al. 2005), molecular hydrogen (Philip et al. 2004; Ubachs & Reinhold 2004) and its isotopologue HD (Ivanov et al. 2008). Using a tunable, narrow-band laser source in the range 90–110 nm, transition wavelengths could be determined with an absolute accuracy of 0.000 008 nm or  $\Delta\lambda/\lambda = 8 \times 10^{-8}$ . In this paper, we extend these studies to the oxygen atom for which we report the accurate wavelength measurement of 16 transitions from the ground state to various excited states of O I. Some of these transitions are contained in the Berengut list (2006).

The XUV absorption spectrum of O I has been obtained by classical means with Doppler broadening present, the first detailed study being that by Huffman, Larrabee & Tanaka (1967a,b). The absolute calibration uncertainties in their measured wavelengths were estimated to be about  $\pm 0.002$  nm or  $\pm 2$  cm<sup>−1</sup>, although the relative uncertainties were considerably less. The most recent atomic data base compilation is that of Morton (1991), for which the O I data are taken from Moore (1976). Inspection of the data in the latter compilation shows that the term values for the final states accessed in our present absorption measurements derive from Eriksson & Isberg (1963, 1968). The tabulated values are theoretical ones, calculated by these authors, and are deemed to be accurate to  $\pm 0.000 05$  nm, but have never been confirmed with experimental observations at that accuracy. With the present laser-based sub-Doppler measurements, the accuracy of these term values is improved considerably. To obtain improved experimental excited-state level energies, the highly accurate data from a far-infrared study by Zink et al. (1991) on the splittings between the fine-structure components in the <sup>3</sup>P ground state of O I are employed. These data are also used to verify the consistency of our results. In a later Letter (De Natale et al. 1993), the ground-state splittings in the oxygen atom, again obtained by employing far-infrared spectroscopy, essentially confirmed the previous findings (Zink et al. 1991) within our stated accuracy.

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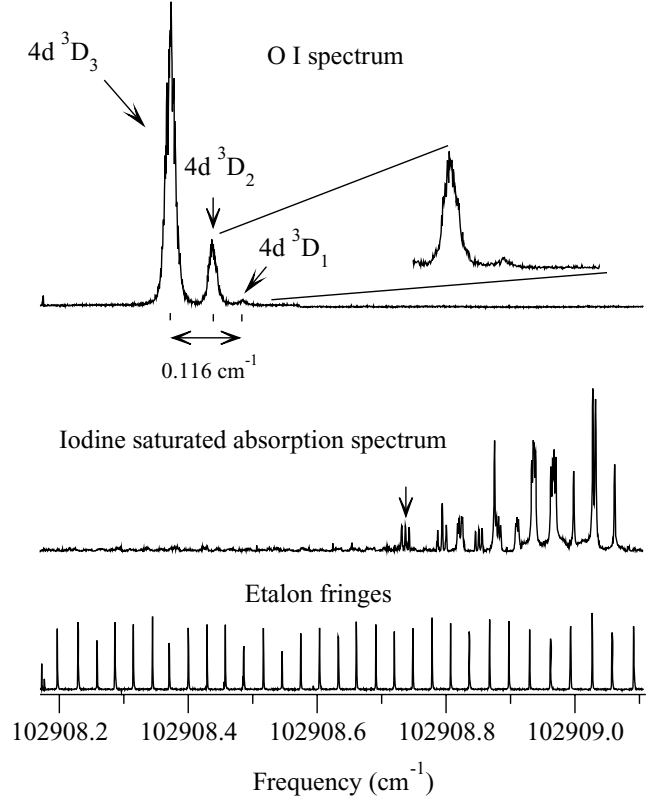
## 2 EXPERIMENTAL

The operation and characteristics of the tunable, narrow-band XUV laser source and its application in spectroscopic studies have been described previously (Ubachs et al. 1997). In short, laser and non-linear optical techniques are combined to generate  $\sim 5$  ns pulses of tunable XUV radiation. A continuous wave (cw) seed beam produced in the visible spectral range, is pulse amplified, frequency doubled in a KD\*P-crystal, and subsequently tripled in frequency in a gas jet to produce coherent radiation with a bandwidth ( $\Delta\nu_{\text{bw}}$ ) of 250 MHz, yielding  $\Delta\lambda_{\text{bw}}/\lambda \sim 10^{-7}$ . Spectroscopic measurements are performed in a crossed-beam configuration, where the collinear XUV and UV laser light is perpendicularly intersected with an atomic beam, providing a largely reduced Doppler width. A pulsed-discharge source based on either pure oxygen or a 1:2 oxygen/helium mixture is used to produce ground-state oxygen atoms in a beam. The procedure is similar to that employed to produce carbon (Labazan et al. 2005) and nitrogen (Salumbides et al. 2005) atoms in their respective ground states. The detection scheme involves resonant excitation of the oxygen atoms, subsequent UV-induced photoionization, followed by mass selection in a time-of-flight (TOF) setup and time-gated detection of the resulting ions. In our experiments only the arrival of  $^{16}\text{O}$  ions is detected. Hence, our spectroscopic results should be assigned to the pure  $^{16}\text{O}$  isotope, rather than to naturally occurring oxygen as is usually the case in the classical absorption experiments.

Frequency or wavelength calibration is performed by referencing the cw-seed radiation against an  $\text{I}_2$  molecular standard (Xu et al. 2000), while also employing an interpolation between frequency markers of a stabilized étalon. A typical example of an observed O I transition together with the  $\text{I}_2$  and étalon marker traces is presented in Fig. 1. The instrumental line width is mainly determined by residual Doppler broadening in the crossed-beam setup, and by the bandwidth of the XUV radiation, yielding a total of  $\sim 500$ – $600$  MHz. The absolute accuracy of the wavelength positions of the O I transitions is determined by a number of factors, where a systematic uncertainty due to the frequency chirp in the dye-amplification system gives the largest contribution (Ubachs et al. 1997). For all observed transitions, we estimate the relative uncertainty to be  $\Delta\lambda/\lambda \sim 8 \times 10^{-8}$ .

## 3 RESULTS AND DISCUSSION

Atomic oxygen is produced in its  $^3\text{P}$  ground state, in each of the well-known fine-structure components  $^3\text{P}_{2,1,0}$ . The  $^3\text{P}_2$  component is lowest in energy, with  $^3\text{P}_1$  and  $^3\text{P}_0$  158.268 741(5) and 226.985 249(8)  $\text{cm}^{-1}$  above it, respectively (Zink et al. 1991). The population of the three fine-structure components depends on the details of the oxygen atom production in the discharge. Since only  $^{16}\text{O}$  (which has a nuclear spin zero) is studied in this work, hyperfine splittings are absent. Because the spin-orbit coupling in atomic oxygen is small, the one-photon selection rules  $\Delta J = 0, \pm 1$ ;  $\Delta S = 0$  and  $\Delta l = \pm 1$  hold. The excited states accessed are O  $(1s)^2(2s)^2(2p)^3(^4\text{S}^\circ)$  3d, 4d and 5d  $^3\text{D}_{3,2,1}^\circ$  with six possible transitions each, O  $(1s)^2(2s)^2(2p)^3(^2\text{D}^\circ)$  3s,  $^3\text{D}_{3,2,1}^\circ$  with six possible transitions and O  $(1s)^2(2s)^2(2p)^3(^4\text{S}^\circ)$  5s and 6s  $^3\text{S}_1^\circ$  with three possible transitions each. Not all possible transitions are observed in this work. The reasons for this are the following: (i) the  $^3\text{P}_0$  and  $^3\text{P}_1$  levels of the ground-state multiplet are probably less populated than  $^3\text{P}_2$ ; (ii) considering the degeneracy factors the transitions originating from  $^3\text{P}_0$  and  $^3\text{P}_1$  are expected to be weaker than those from  $^3\text{P}_2$ ; (iii) the fine-structure splittings in higher Rydberg states decrease, and can no longer be resolved



**Figure 1.** Recording of the  $(^4\text{S}^\circ) ^3\text{P}_2 \rightarrow (^4\text{S}^\circ) 4d ^3\text{D}^\circ$  triplet in atomic oxygen performed via  $1 \text{ XUV} + 1' \text{ UV}$  ionization spectroscopy. The middle and lower traces show the  $\text{I}_2$ -saturation spectrum used for absolute calibration and the markers of an actively stabilized étalon used for relative calibration. The marked iodine line is the  $t$ -hyperfine component of the B–X(18,2) R(109) rotational line in  $\text{I}_2$  at  $17\,151.456\,06 \text{ cm}^{-1}$ .

in the case of the O  $(1s)^2(2s)^2(2p)^3(^4\text{S}^\circ) 5d ^3\text{D}_{3,2,1}^\circ$  level. Since the line widths for the transitions arising from the  $^3\text{P}_1$  and  $^3\text{P}_2$  ground-state fine-structure components are observed to be  $\sim 0.015 \text{ cm}^{-1}$ , this value constitutes an upper limit on these splittings. A total of 16 transitions are observed in this study. Another eight are calculated combining the present measurement with the ground-state data from the literature (Zink et al. 1991). The results are summarized in Table 1.

The measurements were taken during several runs on different days. The statistical errors in a certain run were small and of the order of  $0.0015 \text{ cm}^{-1}$  ( $0.000\,0015 \text{ nm}$ ). However, systematic errors between runs, probably associated with minor day-to-day differences in alignment and chirp, were significantly larger. Hence, on the basis of the error budget, we attribute an uncertainty of  $\pm 0.008 \text{ cm}^{-1}$  ( $\pm 0.000\,008 \text{ nm}$ ) to each transition in Table 1. All wavelengths pertain to vacuum.

The assignments of the various transitions are straightforward and based on literature data (Moore 1976). Since the fine-structure splittings in the  $^3\text{P}$  ground state are known to extremely high accuracy (Zink et al. 1991), they can be used, together with our precise transition frequencies, to determine the energies of the final excited states with unprecedented accuracy. In Table 2, the excited-state energies of the levels studied in this work are summarized. These energies are given with respect to the  $^3\text{P}_2$  ground-state level of the oxygen atom. By considering different combinations of measured transitions and ground-state splittings leading to the same upper state, the

**Table 1.** Observed line positions (in  $\text{cm}^{-1}$  and nm) of transitions originating from the ground-state multiplet of oxygen atoms ( $^{16}\text{O}$  isotope). Since the systematic errors exceed the statistical ones, for all transitions an uncertainty of  $\pm 0.008 \text{ cm}^{-1}$  or  $\pm 0.000008 \text{ nm}$  is estimated.

Initial state	Final state	Frequency ( $\text{cm}^{-1}$ )	Wavelength (nm)
$^3\text{P}_0$	$(^4\text{S}^\circ) 3\text{d } ^3\text{D}_1^\circ$	97 261.383	102.815 729
$^3\text{P}_1$	$(^4\text{S}^\circ) 3\text{d } ^3\text{D}_1^\circ$	97 330.100	102.743 139
$^3\text{P}_1$	$(^4\text{S}^\circ) 3\text{d } ^3\text{D}_2^\circ$	97 330.159	102.743 077
$^3\text{P}_2$	$(^4\text{S}^\circ) 3\text{d } ^3\text{D}_1^\circ$	97 488.369	102.576 339
$^3\text{P}_2$	$(^4\text{S}^\circ) 3\text{d } ^3\text{D}_2^\circ$	97 488.429	102.576 276
$^3\text{P}_2$	$(^4\text{S}^\circ) 3\text{d } ^3\text{D}_3^\circ$	97 488.530	102.576 170
$^3\text{P}_1$	$(^2\text{D}^\circ) 3\text{s } ^3\text{D}_2^\circ$	100 989.248*	99.020 442*
$^3\text{P}_2$	$(^2\text{D}^\circ) 3\text{s } ^3\text{D}_3^\circ$	101 135.394	98.877 352
$^3\text{P}_2$	$(^2\text{D}^\circ) 3\text{s } ^3\text{D}_2^\circ$	101 147.517	98.865 502
$^3\text{P}_0$	$(^4\text{S}^\circ) 5\text{s } ^3\text{S}_1^\circ$	102 184.994*	97.861 727*
$^3\text{P}_1$	$(^4\text{S}^\circ) 5\text{s } ^3\text{S}_1^\circ$	102 253.710*	97.795 962*
$^3\text{P}_2$	$(^4\text{S}^\circ) 5\text{s } ^3\text{S}_1^\circ$	102 411.979	97.644 827
$^3\text{P}_0$	$(^4\text{S}^\circ) 4\text{d } ^3\text{D}_1^\circ$	102 681.512*	97.388 515*
$^3\text{P}_1$	$(^4\text{S}^\circ) 4\text{d } ^3\text{D}_2^\circ$	102 750.181*	97.323 430*
$^3\text{P}_1$	$(^4\text{S}^\circ) 4\text{d } ^3\text{D}_1^\circ$	102 750.229*	97.323 384*
$^3\text{P}_2$	$(^4\text{S}^\circ) 4\text{d } ^3\text{D}_3^\circ$	102 908.382	97.173 814
$^3\text{P}_2$	$(^4\text{S}^\circ) 4\text{d } ^3\text{D}_2^\circ$	102 908.449	97.173 751
$^3\text{P}_2$	$(^4\text{S}^\circ) 4\text{d } ^3\text{D}_1^\circ$	102 908.498	97.173 705
$^3\text{P}_0$	$(^4\text{S}^\circ) 6\text{s } ^3\text{S}_1^\circ$	104 938.226*	95.294 159*
$^3\text{P}_1$	$(^4\text{S}^\circ) 6\text{s } ^3\text{S}_1^\circ$	105 006.943*	95.231 799*
$^3\text{P}_2$	$(^4\text{S}^\circ) 6\text{s } ^3\text{S}_1^\circ$	105 165.211	95.088 479
$^3\text{P}_0$	$(^4\text{S}^\circ) 5\text{d } ^3\text{D}_1^\circ$	105 182.024	95.073 279
$^3\text{P}_1$	$(^4\text{S}^\circ) 5\text{d } ^3\text{D}_{1,2}^\circ$	105 250.733	95.011 214
$^3\text{P}_2$	$(^4\text{S}^\circ) 5\text{d } ^3\text{D}_{1,2,3}^\circ$	105 409.010	94.868 551

\*Calculated using a measured transition and the known ground-state level splittings.

uncertainties in some term energies are reduced. A comparison with the calculated values from Moore's tables shows typical deviations of the order of  $0.02 \text{ cm}^{-1}$  ( $0.00002 \text{ nm}$ ) or less. This supports the claim of Eriksson & Isberg (1968) that their calculated values are accurate to  $\pm 0.00005 \text{ nm}$ .

The chirp effects and possible remaining Doppler shifts represent by far the largest contributions to the total error budget. Since for transitions in narrow wavelength ranges the contributions of these effects are virtually identical, the frequency splittings between excited-state fine-structure components can be determined more accurately. In Table 3, these splittings are presented with respect to the lowest lying component of each multiplet. Since the relevant errors are now statistical rather than systematic, they are shown in brackets in the table.

It should be noted that the multiplet splittings associated with the  $(1\text{s})^2(2\text{s})^2(2\text{p})^3(^4\text{S}^\circ) 3\text{d } ^3\text{D}^\circ$  and  $4\text{d } ^3\text{D}^\circ$  levels have opposite signs. This is probably due to slightly different spin-orbit-mediated interactions with the  $(1\text{s})^2(2\text{s})^2(2\text{p})^3(^4\text{S}^\circ) 3\text{d}$  and  $4\text{d } ^5\text{D}^\circ$  quintuplet levels, located slightly below the corresponding triplets (Moore 1976).

Since our results are exclusively for the  $^{16}\text{O}$  isotope, their relevance for a comparison with quasar data, based on the isotopic composition of atomic oxygen in distant cosmic clouds, should be

**Table 2.** Experimental term energy values for the  $^{16}\text{O}$  isotope of atomic oxygen and comparison with calculated values from Moore's Tables. The uncertainties are given in brackets.

Level	Energy ( $\text{cm}^{-1}$ )	Moore ( $\text{cm}^{-1}$ )
$(^4\text{S}^\circ) 3\text{d } ^3\text{D}_1^\circ$	97 488.368 (3)	97 488.378
$(^4\text{S}^\circ) 3\text{d } ^3\text{D}_2^\circ$	97 488.428 (4)	97 488.448
$(^4\text{S}^\circ) 3\text{d } ^3\text{D}_3^\circ$	97 488.530 (8)	97 488.538
$(^2\text{D}^\circ) 3\text{s } ^3\text{D}_3^\circ$	101 135.394 (8)	101 135.407
$(^2\text{D}^\circ) 3\text{s } ^3\text{D}_2^\circ$	101 147.517 (8)	101 147.526
$(^4\text{S}^\circ) 5\text{s } ^3\text{S}_1^\circ$	102 411.979 (8)	102 411.995
$(^4\text{S}^\circ) 4\text{d } ^3\text{D}_3^\circ$	102 908.382 (8)	102 908.374
$(^4\text{S}^\circ) 4\text{d } ^3\text{D}_2^\circ$	102 908.449 (8)	102 908.443
$(^4\text{S}^\circ) 4\text{d } ^3\text{D}_1^\circ$	102 908.498 (8)	102 908.489
$(^4\text{S}^\circ) 6\text{s } ^3\text{S}_1^\circ$	105 165.211 (8)	105 165.232
$(^4\text{S}^\circ) 5\text{d } ^3\text{D}_{1,2,3}^\circ$	105 409.010 (8)	105 409.008

**Table 3.** Calculated frequency splittings of the excited-state fine-structure components. The uncertainties are shown in brackets.

Level	$J$	Energy ( $\text{cm}^{-1}$ )
$(^4\text{S}^\circ) 3\text{d } ^3\text{D}^\circ$	1	0.0
	2	0.060 (2)
	3	0.161 (2)
$(^2\text{D}^\circ) 3\text{s } ^3\text{D}^\circ$	3	0.0
	2	12.123 (1)
$(^4\text{S}^\circ) 4\text{d } ^3\text{D}^\circ$	1	*
	3	0.0
	2	0.067 (3)
	1	0.116 (3)

\*Transitions to  $(^2\text{D}^\circ) 3\text{s } ^3\text{D}_1^\circ$  have not been observed.

discussed. Hence, the role of the other stable isotopes  $^{17}\text{O}$  and  $^{18}\text{O}$  should be considered. Their possible importance depends on their isotope shifts compared to  $^{16}\text{O}$  and their abundances. For atomic oxygen, there are two main contributions to the isotope shift: the Normal Mass Shift (NMS), also called Bohr's shift, and a Specific Mass Shift (SMS). For a light atom as oxygen the volume shift may be neglected. Hence, for every energy level one obtains (Godefroid et al. 1999):

$$E_M = \frac{M}{(1+M)} E_\infty + \frac{M}{(1+M)^2} S_{\text{SMS}},$$

where  $E_M$  is the energy level corresponding to a finite mass  $M$  (in units of electronic mass  $m_e$ ), and  $E_\infty$  is the (negative) energy eigenvalue of the infinite nuclear mass problem. Also

$$S_{\text{SMS}} = -\langle \Psi_\infty | \sum_{i < j} \nabla_i \cdot \nabla_j | \Psi_\infty \rangle.$$

The effect of the first term (NMS) shifts the levels of the various isotopes by different amounts that can be easily calculated when the excited-state energy is known. For the excited states addressed in this study, these different amounts lie in the range  $0.36\text{--}0.40 \text{ cm}^{-1}$ . The SMS depends on the excited-state wavefunction for the infinite mass problem, and even its sign is not known a priori. However, it can be safely stated that the overall isotope shifts between the various stable isotopes of atomic oxygen for the excited states under consideration will not amount to more than  $0.7 \text{ cm}^{-1}$ .

For a comparison with quasar data, the isotopic abundances in the cosmic clouds should be considered as well.  $^{16}\text{O}$  is a principal

product of stellar evolution, primarily produced at the end of the helium burning in stars, and therefore very abundant in the Universe. In contrast, the production of  $^{17}\text{O}$  and  $^{18}\text{O}$  secondary isotopes requires pre-existing seed nuclei. Hence, the abundances of  $^{17}\text{O}$  and  $^{18}\text{O}$  are low, with terrestrial abundances of typically 0.0372(4) and 0.200004(5) per cent (Rosman & Taylor 1998), respectively. Employing these values and assuming a realistic line width for the astronomical measurements to be  $\sim 1 \text{ cm}^{-1}$  (taken on the XUV scale, hence for a resolving power of 100 000), we performed simulations for a range of isotope shifts to determine whether the simultaneous observation of all isotopes would shift the line position of a natural O-sample from the presently determined lines for  $^{16}\text{O}$ . Our results show a maximum deviation of  $0.0002 \text{ cm}^{-1}$ , negligible compared to the uncertainties obtained with telescope systems currently in use for high-redshift observations (HIRES-Keck and UVES-VLT). A comparison with our  $^{16}\text{O}$  results is therefore valid.

#### 4 CONCLUSION

Experimental transition energies to highly excited states in atomic oxygen were obtained with a tunable narrow-band XUV laser source at an unprecedented accuracy of  $\Delta\lambda/\lambda \sim 8 \times 10^{-8}$ . A comparison with previously calculated term values of the excited states accessed in the present experiments shows deviations of up to  $0.02 \text{ cm}^{-1}$  (0.00002 nm). The present values can be used in comparisons with astrophysically observed O I transitions, in order to establish a possible variation in the fine-structure constant  $\alpha$  over a time-span of  $\sim 10^{10}$  yr.

#### ACKNOWLEDGMENTS

The authors thank the Netherlands Foundation for Fundamental Research on Matter (FOM) for financial support.

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